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#### **EUROPEAN PATENT APPLICATION**

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(54) Gaseous diffusion resistant article and method utilizing same.

of a liquid resistant plastics material, and an outer layer (5) of a plastics impact-resistant material, and has an intermediate layer (3) of a gaseous diffusion resistant plastics material interposed between and bonded to the core and the outer layer. The outer layer and the core are preferably made of a cross-linked polyolefinic material such as cross-linked polyothylene. The intermediate layer is preferably selected such that it is an adhesive for both the core and outer layer materials and such that it has a measure of elasticity to compensate the relative coefficients of linear expansion between the intermediate layer and its adjoining layers.

Tubes in accord with this invention may be formed by co-extrusion with economy and precision and are particularly useful in central heating applications.

#### DESCRIPTION

The present invention relates to a gaseous diffusion resistant tube comprising a core of a liquid-resistant plastics material and an outer layer of a plastics impact-resistant material. Particularly it relates to such an arrangement for use in hot liquid transfer pipes such as those utilized in central heating systems.

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Plastics pipes, particularly those of cross-linked polyethylene are widely used for the transfer of hot liquids; particularly heat transfer liquids for use in central heating systems. At the temperatures at which the central heating systems operate, oxygen from the air particularly, diffuses from the exterior of said pipe into the water or other heat exchange liquid in the pipe. The oxygen borne by the liquid in the pipe contacts exposed metal surfaces in the system and tends to oxidise the same

metals then become detached and are transferred, suspended in the heat exchange liquid, to a constriction or other obstruction where they are deposited and can eventually cause blockages. Thus, it is generally desirable to provide oxygen diffusion resistant tubes for use in central heating systems, but for various reasons those currently available are not wholly satisfactory.

It is known, for example from British Patent

10 No: 1,175,005, to provide a two-layer gaseous diffusion resistant tube made, for example, from polyethylene and nylon. Such an arrangement is not suitable for central heating installations however because of its linear expansion during heat cycling. Where the polyethylene is cross-linked to provide suitable dimensional stability said oxygen resistant nylon coating overlying the same cannot be used successfully because its coefficient of linear expansion is so different from that of the cross-linked polyethylene tube.

Similarly, it has been proposed, for example in

British Patent No: 1,171,122 to provide a gaseous diffusion
resistant tube having an intermediate layer of a metallic
foil which both reinforces the tube and prevents gaseous
diffusion. Such assemblies are difficult and hence

expensive to produce mainly because of the difficulty of bonding the exterior and interior plastics layers to the foil. Further the installation of such tubes is difficult because of their inherent rigidity and because plastics end fittings utilized in central heating systems are not generally suitable for use with pipes including a metal insert.

The present invention seeks to provide therefore a flexible gaseous diffusion resistant tube which is 10 usable under heat cycling conditions, which can be co-extruded, and is easy to instal.

According to the present invention, therefore, there is provided a gaseous diffusion resistant tube comprising a core of a liquid resistant plastics material, and an outer layer of a plastics impact-resistant material, characterised by an intermediate layer of a gaseous diffusion resistant plastics material interposed between and bonded to the core and the outer layer. It is preferred that the core and the outer layer have substantially similar coefficient of linear expansion and the intermediate layer is slightly elastic and adhesive to both the core and the outer layer. The core and/or the outer layer may be made of a cross-linked polyolefinic material such as cross-linked polyethylene which much improves the

Materials for the intermediate layer such as polyvinylalcohol (PVAL) are particularly useful in the practice of this invention, but are not, in themselves, impact-resistant and hence must be protected by an impact resistant layer. The impact-resistant layer in such circumstances need have no diffusion resistant qualities and hence, if intrinsically strong, may be applied as a comparatively thin layer.

Polyethylene in linear or cross-linked polymeric

10 form is an example of such an impact-resistant layer.

Another example of a particularly desirable gaseous

diffusion resistant material is polyacrylonitrile (PAN).

Other materials which may be used as the impact-resistant

layer include copolymers of acrylonitrile and thermo
plastics polyesters, polyamides or chlorinated PVC.

The particular gaseous diffusion resistant layer for incorporation with the plastics material of the core will in general be selected dependent upon the atmosphere in which the eventual pipe is to be used. Thus, in a normal central heating system it is most important to prevent the diffusion of oxygen into the heat exchange liquid.

However, in certain laboratory applications the gaseous diffusion resistant material is sel cted to inhibit the diffusion of halogens such as chlorine or other organic

Some materials such as PVAL when used in the gaseous diffusion-resistant layer also act as an adhesive and can be utilized, as stated above, to adhere the core to the outer impact-resistant layer selected.

However, when the impact-resistant layer or the exterior surface of the core does not readily adhere to the gaseous diffusion-resistant layer a separate adhesive can be used.

Where possible it is desirable to coaxially co
extrude sequentially the core, the diffusion-resistant
layer, and the impact-resistant layer. In some applications
it is convenient to utilize the same material, eg:

cross-linked polyethylene as both the impact
resistant layer and the core. This ensures that the

coefficience of linear expansion of the core and the
outer layer are similar; where PVALfor example is used
as the intermediate layer its slight elasticity compensates
for its slightly different coefficient of linear expansion.

Some embodiments of the invention will now be described

20 by way of illustration only and with reference to the
following examples and drawings.

In the accompanying drawings:-

Figures 1 and 2 represent diagrammatic cross-sections
25 through segments of oxygen diffusion resistant tubes of the invention.

Figure 1 shows a cross-linked polyethylene (PEX) tube 1 fused to an oxygen diffusion resistant layer of PVAL3 hich in turn is fused to an impact resistant layer of linear polyethylene per se 5; and

Figure 2 shows a PEX tube 1 having an adhesive interlayer 2' and 4' disposed respectively between the PEX tube 1 and an oxygen diffusion resistant layer PAN 3 and between the said layer 3 and a polyethylene impact resistant layer 5.

Where the materials forming layers 3 and 5 cannot be coextruded with the tube 1 they may be applied by spraying, dipping or wrapping as required. The thickness of the layer 3 should be such as to reduce oxygen diffusion by at least 1/5th of that of the core material, and it is preferred that the thickness of the layer 3 is between 1/100 and 1/10 of the wall thickness of the core.

By way of this example, the following layers were all applied to samples of a PEX tube. Said tube had an outer diameter of 20 mm and an inner wall thickness of 2 mm. Each sample was tested for oxygen diffusion against a similar control sample which had either no coating thereon or only a thin coating of an adhesive with no oxygen diffusion resistant capabilities.

The results are set forth in the following table:

### TABLE 1

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Impact and oxygen diffusion resistant samples with and without separate impact resistant layer.

		RIAL OF SFER PIPE	OXYGEN DIFFUSION RESISTANT LAYER	SEPARATE IMPACT RESISTANT LAYER	COMPARATIVE DIFFUSION CHARACTERISTICS
10	1)	2mm PEX	20 µ PVAL	0.3 mm PE	< 1/5th control
	2)	2mm PEX	20 / PAN	0.3 mm PE	< 1/5th control
	3)	2mm PEX	40 M PVAL	0.3 mm PE	< 1/10th control
	4)	2mm PEX	40 / PAN copolymer BAZEX (RTM)	0.3 mm PE	<1/10th control
15	5)	2mm PEX	40 chlorinated PVC.	0.3 mm PE	< 1/5th control

It can thus be seen that particularly advantageous results can be achieved with a layer between 20 and 40 \( \mu\) thick of PVAL or PAN over a PEX tube. This will reduce oxygem diffusion 20 to less than 1/10th. of the diffusion expected through a PEX tube with a 2 mm wall thickness and an outer diameter of 20 mm.

The use of tubes made in accord with the invention has been found in practice to considerably reduce the

25 instance of corrosion in central heating systems and this

leads to a reduction in the required servicing frequencies.

Because the diffusion resistant layers can be formed during the production of normal PEX tubes, tubes according to the invention can be produced for much the same cost

as that of ordinary PEX tubes. Consequently, oxygen diffusion resistant pipes can be utilized in the installation of central heating systems without a significant extra cost and with material long term benefits. It will also be appreciated that the comparatively thin non-structural oxygen diffusion layer allows of a greater flexibility than with metal foil oxygen diffusion resistant layers and allows a close match of coefficients of linear expansion by virtue of the slight elasticity of said oxygen diffusion resistant layer.

15 Tubes of this invention have a materially improved long term structural integrity when utilized in central heating installations.

#### CLAIMS

- 1. A gaseous diffusion resistant tube comprising a core of a liquid resistant plastics material and an outer layer of a plastics impact resistant material, characterised by an intermediate layer of a gaseous diffusion resistant plastics material interposed between and bonded to the core and the outer layer.
- A tube according to claim 1 wherein the core and the outer layer have substantially similar coefficients of
   linear expansion and the intermediate layer has at least a slight elasticity.
  - 3. A tube according to claim 1 or claim 2 wherein the core at least is made of a cross-linked polyolefinic material.

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- 4. A tube according to claim 3 wherein the outer layer is also made of a polyolefinic material.
- 5. A tube according to either of claims 3 or 4 wherein 20 the cross-linked polyolefinic material is cross-linked polyethylene.
  - 6. A tube according to any one of the preceding claims wherein the intermediate layer is also adhesive to the

material forming the core and the material forming the outer layer.

- 7. A tube according to claim 6 wherein the intermediate
  5 layer is selected from polyvinylalcohol, polyacrylonitrile
  or polyvinylidene chloride.
  - 8. A tube according to any preceding claim wherein the intermediate layer has a thickness between 60 \text{M} and 10 \text{M}.
- any one of claims 1 to 8 which comprises sequentially forming the core layer, the intermediate layer and the outer impact resistant layer in coarial array such that said layers are fused together at their interfaces.

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- 10. A method according to claim 9 wherein the forming step is effected by coextrusion.
- 11. A central heating system incorporating a tube as
  20 claimed in any one of claims 1 to 8 or made by the method
  of either of claims 9 or 10.





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# **EUROPEAN SEARCH REPORT**

EP 80 30 4008

Cotos	DOCUMENTS CONSIDERED TO BE RELEVAN		CLASSIFICATION OF THE APPLICATION (Int. CL.)
Category	Citation of document with indication, where appropriate, of relevan passages	t Relevant to claim	AT FLICATION (Int. Cl. <sup>3</sup> )
X	US - A - 3 561 493 (MAILLARD)		F 10 L 9/12
	* Column 1, line 57 to column 2,	1-7;9	
	line 71; figures 1,2 *		
	-me video index		
X	US - A - 3 854 504 (MORRISON)		
	* Column 3, lines 49-68; column 4,	1,6	
	lines 30-33; figure 2 *		
7.	The first last		
X	GB - A - 897 222 (FARBWERKE HOECHST)	1-7,9	
	* Page 1, lines 66-73; page 2,		
	lines 24-29; claims 5,7,8; figure 1 *		TECHNICAL FIELDS SEARCHED (Int. Ct. 3)
			F 16 L
x	DE - A - 2 236 375 (TECHNOFORM)	1 2	B 29 F
	* Page 2, lines 4-10; figure *	1,3	
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			CATEGORY OF
			CITED DOCUMENTS
. ]			X: particularly relevant A: technological background
		,	O: non-written disclosure
			P: intermediate document
			T: theory or principle underlying the invention
.			E: conflicting application
			D: document cited in the
			application  L: citation for other reasons
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	The present search report has been drawn up for all claims	member of the same patent family,	
of sear	,	Examiner	corresponding accument
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